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REPORT SUMMARY

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ADVANCED RESEARCH PROJECTS AGENCY

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NAME OF CONTRACTOR:

The University of Rochester
College of Engineering & Applied Science
Rochester, New York

TITLE OF PROJECT:

6 Stimulated Soft X-Ray Emission in the
Range of 40A to 140A from Laser Excited
Media.

CO-PRINCIPAL INVESTIGATORS:

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A program to obtain experimental evidence for soft x-ray laser action is described. Analysis of the pumping requirements for an x-ray laser point to the use of a high density medium, probably having a high temperature, as a soft x-ray amplifier. We concentrated our study on determining the suitability of a laser produced plasma for this purpose. Measurements of the relevant plasma parameters (electron temperature and density, plasma scale length, etc.) were carried out. A comparison of the results with various published proposals for producing soft x-ray laser action in a plasma was made. A scheme involving the use of $3p \rightarrow 3s$ transitions in carbon-like ions was selected for a more detailed study. A computer simulation of a laser plasma was performed, using the HOT SPOT model. The results show that readily measurable gain could be achieved in the wavelength range of 600-800A under the conditions present in our experiment. Relatively straightforward scaling of the results to shorter wavelengths would be possible.

An experiment was designed and partially assembled to test the results of the computer simulation. The ARPA program was terminated before the experiment could be carried out.

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 Washington, D.C. 20375
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I. INTRODUCTION

The first report of maser action was made in 1953⁽¹⁾ and laser action at visible wavelengths was first reported in 1959.⁽²⁾ Since that time progress towards observing laser action at shorter wavelengths has been slow and at this writing there is still no evidence for direct laser action below 1000Å. Only recently have conventional laser sources been upconverted to wavelengths below 1000Å.⁽³⁾ The difficulties in producing laser action below 1000Å have been sufficient for many workers to consider this the wavelength boundary of the x-ray region and for the phrase "1000Å barrier" to be coined in this connection.⁽⁴⁾

Few naturally occurring or man-made materials exhibit chemical binding energies in excess of 10ev or ionization potentials in excess of 20ev. Thus we face a formidable materials problem in seeking to pump a transition to inversion whose energy exceeds 12ev (1000Å) since it is reasonable to expect a significant fraction of the total pump energy to be deposited in the environment of the laser medium. Changes induced in the characteristics of the pumped medium could preclude maintenance of species having the desired laser transitions. Thus, to achieve useful amplification it may be important to insist on a medium which is physically stable during the pumping cycle.

Assuming that we can find a stable amplifying medium we then ask what the pumping requirements are for useful gain. If we require oscillation in an optical cavity then we must supply sufficient gain to overcome cavity losses. Neglecting diffraction we require

$$Re^{gL} \geq 1 \quad (1)$$

where R is the mirror reflectance in the cavity, L is the length of the amplifying medium and g is the gain coefficient,

$$g = \frac{c^2}{4\pi \nu^2} \frac{A}{\Delta\nu} \Delta N \quad (2)$$

In this expression ΔN is the density of inverted species, A is the Einstein coefficient for spontaneous emission and $\Delta\nu$ is the (broadened) linewidth of the laser transition of frequency ν , and c is the velocity of light.

For energetic photons, particularly with energy of the order of 100ev or more, it is not possible to produce mirrors of high reflectance at normal incidence at the current state-of-the-art. If we resort to grazing incidence reflections the loss per surface is lowered but a re-enterant cavity then requires a large number of surfaces. The total round trip loss may not be greatly improved. Therefore, it is helpful to consider the properties of a laser which has no feedback.

If we consider a filament-shaped amplifying medium, then the radiation along the filament axis will become predominantly stimulated when the inversion density exceeds the value⁽⁵⁾

$$\Delta N = \frac{c^2}{8\pi \lambda b} \frac{A}{\Delta\nu} \quad (3)$$

where λ is the transition wavelength and b is the ratio for total rate of decay of the upper laser level to spontaneous radiative decay on the laser transition. The total upper level lifetime, τ , is

$$\tau = \frac{b}{A} \quad (4)$$

This generally implies that high inversion densities will be required at soft x-ray wavelengths because x-ray excited states tend to be short lived, i.e., exciting a forbidden transition is not beneficial if the branching ratio to other decay paths is unfavorable.

To avoid the energy investment required to pump a ground state transition and to minimize the effects of alternate decay channels, most

proposals for direct generation of soft x-ray laser action rely on the use of dipole-allowed excited state transitions. The approximate value of the threshold inversion density vs. transition wavelength for dipole-allowed laser transitions is given in Fig. 1 assuming a branching ratio of unity (the ideal case). If we consider very short wavelengths (say 1\AA) the density of inverted species required becomes an appreciable fraction of solid density and since one is supplying several Kev of energy per excited atom or ion, one is inevitably dealing with a very high energy density environment.

It should be noted that supplying an optical cavity which has significant round trip loss will not alter this conclusion. Equations (1) and (2) can be solved to give the inversion density required to achieve oscillation as a function of the effective reflectivity of the cavity. If this result is compared with the threshold requirement for amplified spontaneous emission (ASE) in equation (3) we may compute the factor by which the inversion density in equation (3) will be reduced by the presence of an optical cavity. In Fig. 2 we plot the factor by which the threshold changes as a function of effective cavity reflectance. It does not appear that proposed optical cavity designs will significantly alter our conclusions as to the requirements for a high energy density environment in producing soft x-ray laser action.

These considerations have led us to suppose that direct generation of laser action below 1000\AA will involve some kind of plasma medium. An important goal in the ARPA program was to achieve laser action at short wavelengths, say 50\AA or less. Only a few plasma devices would appear suitable for producing required environmental temperatures in the 0.1 to 1.0 Kev range at densities approaching solid. During the course of the ARPA program we concentrated our study on the suitability of a laser pro-

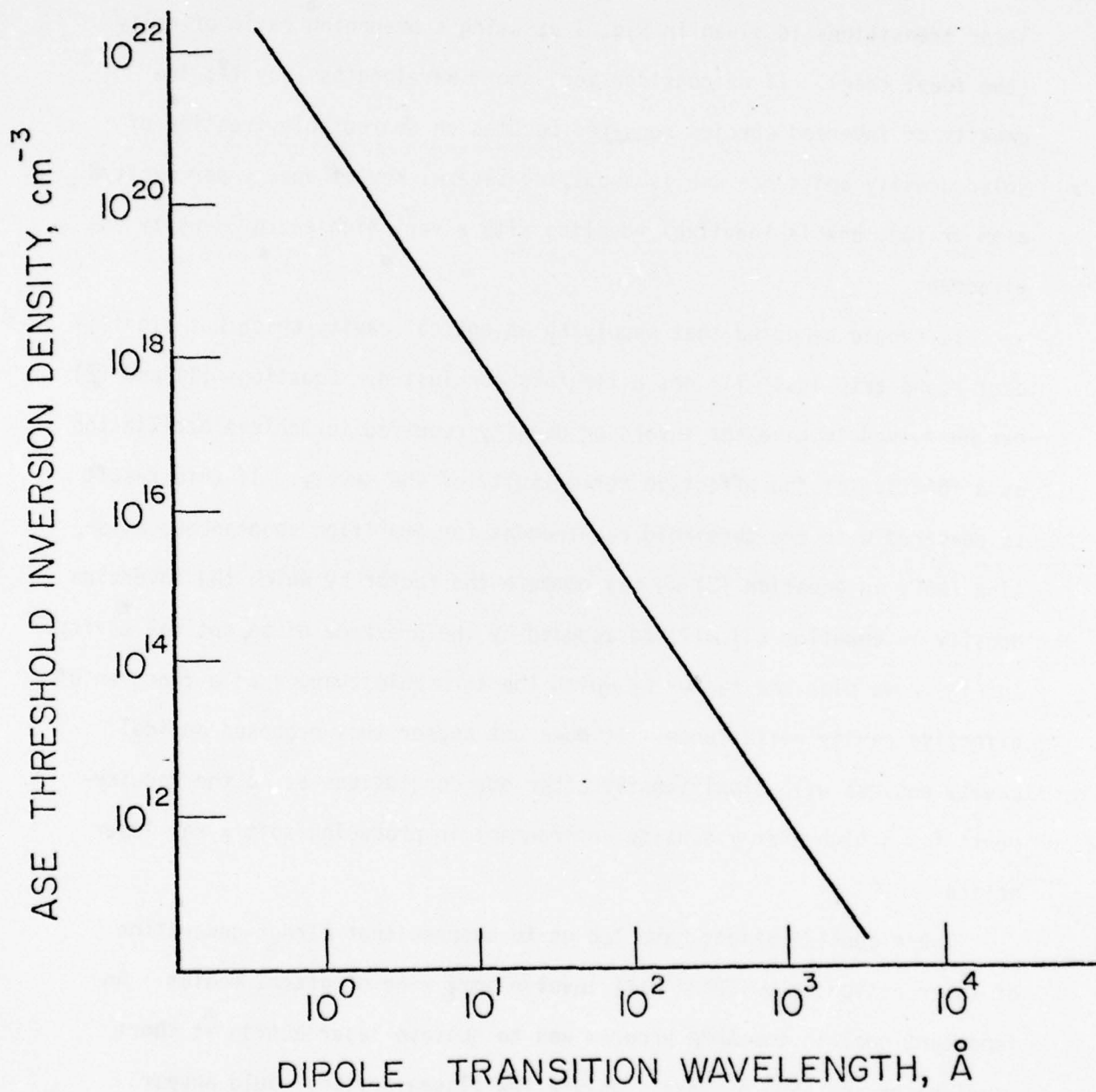


Fig. 1 Approximate ASE threshold inversion density vs. transition wavelength for dipole-allowed excited state transitions.

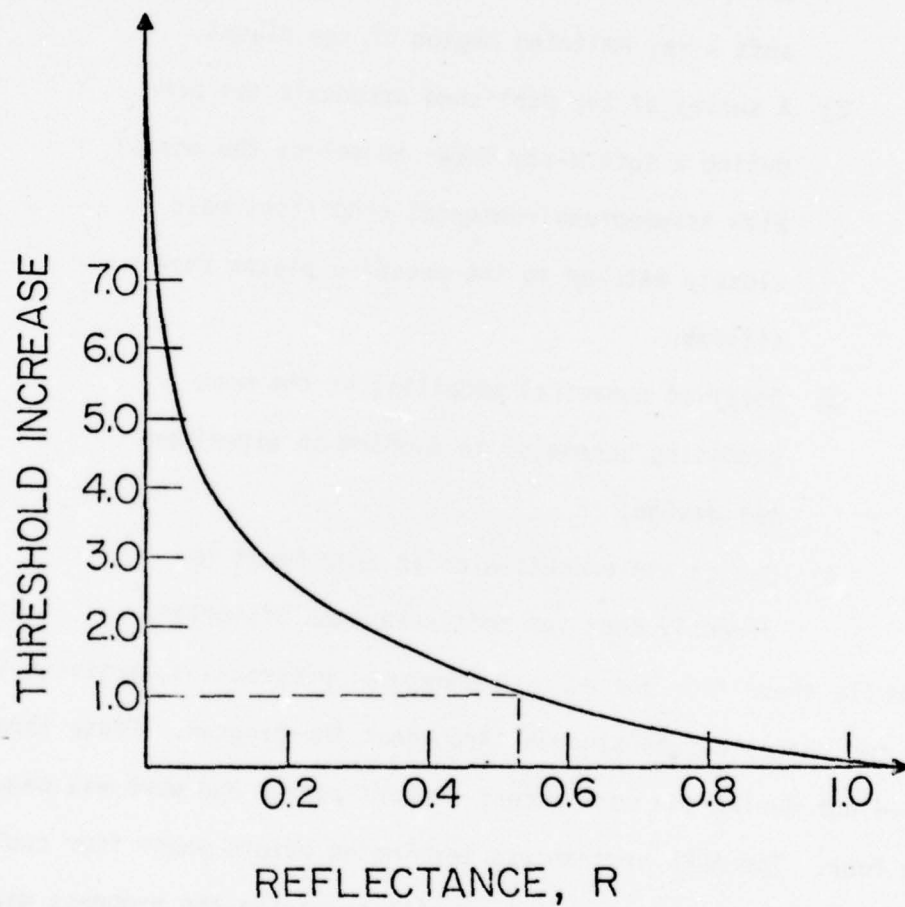


Fig. 2 ASE threshold inversion factor vs. effective cavity reflectance. For net reflectance values below 52% oscillation is more difficult to achieve than is ASE operation.

duced plasma to demonstrate useful amplification at wavelengths down to 50Å or below.

Our program was divided into roughly four phases of activity:

- 1) A detailed experimental study of laser plasma characteristics with emphasis on the active, soft x-ray emitting region of the plasma.
- 2) A survey of the published proposals for producing a soft x-ray laser to select the one(s) with assumed environmental conditions most closely matched to the measured plasma conditions.
- 3) Detailed numerical modelling of the most promising scheme(s) to confirm an experimental design.
- 4) Design and execution of an experiment to directly test for soft x-ray amplification.

While these four phases form a natural progression, activity in the first two phases was continuous throughout the program. Phase three was carried out during the most recent support period and work was begun on phase four. The ARPA program was terminated before phase four could be completed. In this final report we will summarize the progress made in each of these areas up until the program termination date and list the remaining steps required for completion of the study. Reprints of papers published during the most recent support period are attached in the Appendix.

II. EXPERIMENTAL PROGRAM

The most important parameters to be measured in a laser plasma for the purpose of understanding x-ray excited state production are the electron temperature and electron density. The electron temperature determines the approximate degree of ionization of the plasma as well as the energetic limits of accessible excited states. The electron density determines, in part, the density of excited states to be expected; in the case of a plasma with an inverted population it will determine the gain directly. Of equal importance for designing and interpreting a definitive laser experiment are a knowledge of the time history and the spatial distribution of the above parameters.

During the initial support periods we concentrated primarily on the development of instrumentation and analytical tools for the diagnosis of the physical conditions present in a plasma produced by a high peak power, focussed, subnanosecond pulse of radiation from a Nd^{+3} : glass laser.⁽⁶⁾ Results of the activity were given in the two previous technical reports. These results were necessarily fragmentary and thus insufficient upon which to base a realistic experimental design of an x-ray amplifier.

During the last support period a continuation of the laser diagnostics program has enabled us to obtain a more complete understanding of the plasma environment. In this period measurements of the electron temperature and electron density in the plasma were combined with measurements of the plasma density scale length and of the dynamic properties of the active emitting region of the plasma. These measurements, and results of other work published during the support period, form a basis for a geometrical design of a soft x-ray amplifier. The measured plasma conditions also form the basis for the selection of a promising plasma system for the amplifier experiment. (A summary of the numerical modelling of this system is given in the next section).

The measurements summarized here were made with the aid of two experimental facilities which are part of the University of Rochester's Laboratory for Laser Energetics. In one facility a single beam Nd^{+3} : glass laser produces single pulses approximately 200 psec FWHM with up to 20 joules at a wavelength of $1.06\mu\text{m}$. The laser consists of a mode-locked oscillator, pulse switching network and a five stage amplifier system and includes isolation from reflected pulses from the target chamber. This laser system was dedicated exclusively to x-ray laser development and the concurrent development of laser plasma x-ray diagnostic instrumentation. Experiments were performed with the single beam laser in a 24-inch diameter vacuum chamber using a 3.5-inch diameter $f/3.5$ aspheric lens (designed and fabricated at U of R) to focus the laser pulse onto slab targets. Lenses having both circular and cylindrical focal spots are available. Circular focal geometry was employed in diagnostic experiments.

In the second facility a Nd^{+3} : glass laser produces four separate beams for simultaneous irradiation of spherical targets. Each beam is derived from a common oscillator/amplifier chain and delivered up to 50 joules (per beam) at pulsewidths of 250 psec during the experiments described below. (Pulse energies are approximately proportional to pulsewidths over a considerable range, (30-800 psec), and may be varied to suit the experiment.) This system is used primarily for experiments in laser induced fusion. The sophisticated diagnostics available in many of these experiments have yielded important additional information bearing on the design of an x-ray laser experiment. This is direct evidence of the beneficial effect of the strong technology overlap between laser-induced fusion research and the laser plasma approach to x-ray laser development.

A summary of relevant results obtained from the combined facilities will now be given. Time integrated measurements of the plasma electron

temperature and density were made using the single beam facility while the temporal history and spatial distribution measurements were made using the four beam facility. The techniques used to infer the plasma electron temperature and electron density all rely on making a set of assumptions about the relative importance of various physical processes. To improve our confidence in these techniques we have measured both temperature and density by several methods which emphasize the various assumptions differently. In addition, the measurements have been compared to the results of a detailed numerical simulation of the laser plasma.

The plasma electron temperature may be measured by recording the distribution of the x-ray continuum⁽⁷⁾ (bremsstrahlung) or by the distribution of intensities in the discrete line emission.⁽⁸⁾ Comparison of these measurements with analytic models is facilitated when the plasma is in a high degree of ionization, i.e., when the ions are primarily hydrogen-like and helium-like species. Under our experimental conditions this is true for low to moderate weight elements, i.e., $Z \leq 20$. Accordingly, we concentrated our study in this range and, in particular, to the spectrum of aluminum and silicon plasmas. This choice was made to enable data acquired in connection with laser-induced fusion experiments being conducted at Rochester⁽⁹⁾ to be compared to ours directly. (Laser fusion targets often consist of thin wall glass microballoons with a high pressure gas fill.⁽¹⁰⁾ Silicon ions make a strong contribution to the x-ray line radiation from such targets.)

Both the line and continuum measurements yield a plasma electron temperature between 0.5 and 1.0 Kev depending, of course, on the detailed characteristics of the laser pulse and the target geometry. Over the range of experimental conditions investigated (single beam focused pulse energies between 3 and 15 joules delivered to aluminum slab targets in

approximately 200 psec) the electron temperature is not found to be extremely sensitive to experimental conditions. This has an important bearing on soft x-ray amplifier design -- we will return to this point later.

The plasma electron density has been derived from various features of the distribution of line radiation. The ratios of certain line intensities are density sensitive and provide one kind of measure.⁽¹¹⁾ The observation of Stark broadening from resonance lines from high lying principal quantum levels of the plasma ions yields a second estimate.⁽¹²⁾ These methods indicate that the electron density in the emitting region lies between 10^{20} cm^{-3} and $5 \times 10^{21} \text{ cm}^{-3}$.

More recently we have taken spatially resolved x-ray spectra from four beam irradiation of spherical glass shell targets. In this experiment four laser pulses are focused simultaneously onto the target along axes which lie symmetrically in a plane. A flat crystal spectrometer with an entrance slit oriented in the plane of incidence is used to produce one dimensional, dispersed images of the plasma, viewed at 90° to the plane of irradiation.

The plasma critical surface is located by a characteristic emission of the second harmonic of the incident laser light.⁽¹³⁾ A photograph of the plasma at this wavelength is made, again at 90° to the plane of irradiation. A comparison of the two images is shown in Fig. 3, in which the x-ray image was obtained from resonance line radiation from Na^{+9} .⁽¹⁴⁾ (The sodium is present as a constituent of the glass in the target walls.) Since the plasma density is decreasing radially outward, it appears that the x-ray line emission here is actually produced from a region slightly above critical density. Such an estimate is in good agreement with detailed numerical simulations of the plasma conditions.^(15,16)

As we will show, the above plasma conditions may be favorable to producing inverted populations on transitions in a certain class of plasma ions.

In order for such a population inversion to be readily diagnosed and to enable a practical amplifier geometry to be engineered, it is of great interest to know the geometry and dynamic behavior of the active plasma region. Recent experiments performed in conjunction with research on laser induced fusion at Rochester have provided direct answers to these questions.

It was observed above that the plasma critical surface can be located by photographing the plasma at the second harmonic of the incident laser frequency. A strong scattered field is also emitted by the plasma at $3/2$ the incident laser frequency. This radiation arises through a non-linear interaction between the incident laser field and plasma oscillations occurring at an electron density which is one quarter of the critical value.⁽¹⁷⁾ Thus a photograph of the plasma at $3/2$ the laser frequency serves to locate the region of quarter critical density. A comparison of the two photographs enables a scale length of the density profile to be established.

In our experiments a glass microballoon target approximately $100\mu\text{m}$ in diameter was irradiated by four simultaneous focused laser pulses arranged symmetrically in a plane. The radiation at the two characteristic frequencies was viewed along an axis through the target at 90° to the plane of illumination. Separate images at the two harmonic frequencies were formed on the entrance slit of a high speed electro-optic streak camera.⁽¹⁸⁾ Thus the measurement of the time history of plasma evolution could be studied.

The result of one such experiment is shown in Fig. 4. In this experiment a spherical shell of glass $80\mu\text{m}$ in diameter and having a wall thickness of $1\mu\text{m}$ was irradiated by four beams each having an energy of approximately 40 joules and a pulsewidth of 360 psec FWHM. Micro-densitometer scans of the recorded images were used to determine the positions of the critical and quarter critical points with respect to the target surface. (The

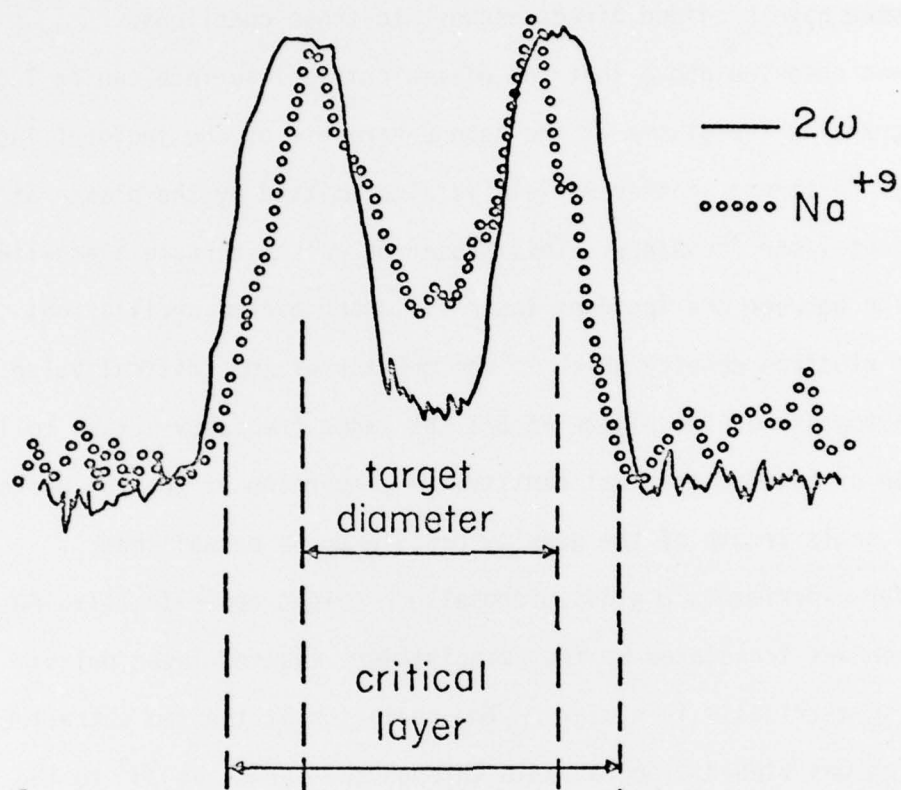


Fig. 3 One-dimensional images of a plasma produced by four simultaneous laser pulses focussed onto a glass microballoon target. The images were photographed at 90° to the plane of illumination and were taken at the second harmonic of the laser frequency (solid curve) and at a soft x-ray emission line of helium-like sodium ions (dotted curve).

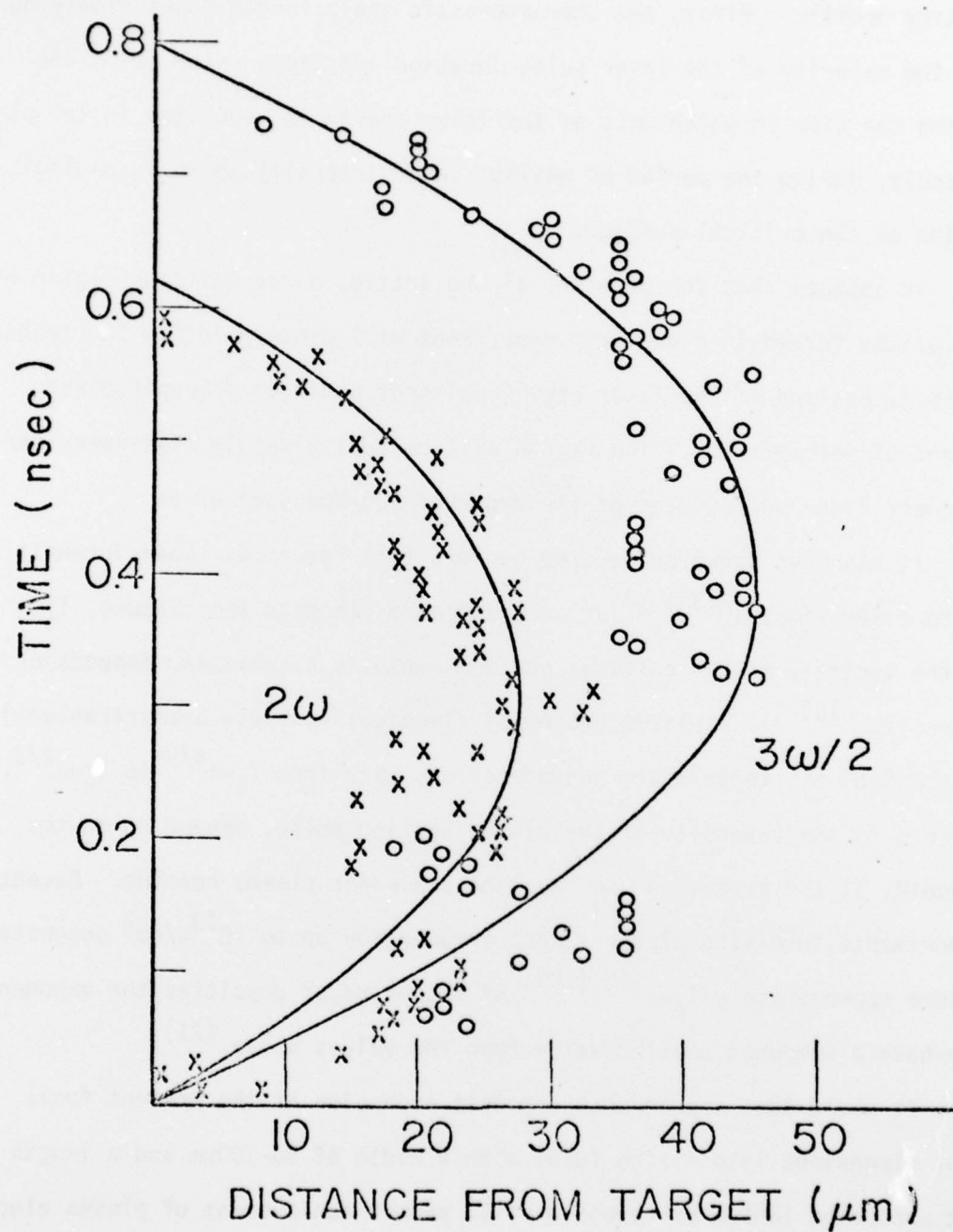


Fig. 4 Time history of the image motion of a plasma produced by four simultaneous laser pulses focussed onto a glass microballoon target. The images were photographed at 90° to the plane of illumination at the second harmonic and $3/2$ harmonic of the laser frequency.

points shown in Fig. 4 are obtained from various scans across a single pair of images.)

Two important conclusions can be drawn from Fig.4 with respect to the density profile. First, the characteristic scale length grows slowly during the majority of the laser pulse duration and has a value below 25μ during the time in which most of the laser energy is deposited in the plasma. Secondly, during the period of maximum laser intensity there is negligible motion of the critical surface.

It appears that the geometry of the active, x-ray emitting region of the plasma formed in a one beam experiment will closely follow the transverse dimensions of the laser beam focal spot and have a longitudinal extent of perhaps $20\mu\text{m}$. The region will be approximately stationary for incident laser pulsewidths of the order of 200-300 psec or so.

It has been shown by several workers that for focal plane intensities up to a few times $10^{13}\text{W}/\text{cm}^2$ or so the plasma electron temperature, T_e , in the vicinity of the critical surface exhibits a sublinear dependence on intensity.⁽¹⁹⁾ In a plasma heated by classical (inverse bremsstrahlung) absorption, the temperature dependence may vary from $T_e \propto \phi^{4/9}$ to $T_e \propto \phi^{2/3}$, where ϕ is the intensity of the plasma heating pulse, depending on the geometry of the expansion and the time scale for plasma heating. Recent experiments involving planar target irradiation up to $10^{13}\text{W}/\text{cm}^2$ suggests a form approximately $T_e \propto \phi^{1/2}$.⁽²⁰⁾ At higher power densities the exponent may have a somewhat smaller value than the values above.⁽²¹⁾

It would thus appear that a modest expansion of the present focal spot dimensions into a line focus with a width of $50\text{-}100\mu\text{m}$ and a length not exceeding $1000\mu\text{m}$ or so would still permit achievement of plasma electron temperatures near 0.5 Kev with presently available laser pulses. A

plasma volume element based on such dimensions has been incorporated into the analysis presented in the next section.

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III. MODELLING AND EXPERIMENTAL DESIGN

The above characterization of plasma conditions allows us to model and design an experiment to verify the presence of an amplifying condition in the plasma. Numerous spectroscopic systems have been suggested for producing inverted populations in a laser excited plasma.^(6,22) These proposals virtually all suggest the use of a plasma region of moderate electron density (less than 10^{20}cm^{-3} , say) either to favor recombination processes, or to avoid collisional mixing of the excited states with a consequent destruction of the inversion.

Calculations based on proposals for recombination pumped inversions generally indicate such inversions to be short lived compared to the laser pulsewidths we have at our disposal.⁽²³⁾ It could prove difficult to detect enhanced spontaneous emission against a large incoherent background. A published experimental study of a recombining plasma suggests a very low density of inverted population may be present, too low to produce useful gain.⁽²⁴⁾ Accordingly, we have re-examined proposals for direct collisional excitation under conditions of high electron density.

The extrapolation of $3p \rightarrow 3s$ transitions in carbon-like ions to the extreme vacuum ultraviolet was analyzed by Elton.⁽²⁵⁾ An appealing feature of this scheme is that it has produced laser action in Z-pinch plasma devices⁽²⁶⁾ and is a CW laser scheme if steady state plasma conditions can be maintained. In Elton's original discussion a rather low electron density was assumed because it was felt that high electron densities would promote collisional mixing of the laser transition and destroy the inversion. However, when the transition energy is smaller than the electron temperature, as in the present case, the rate of electron collisional transition between the levels is roughly the same in both

directions. Considerable time is required to redistribute the excited state populations to steady state values. Plasmas which are produced and heated on timescales of 10^{-10} sec may not exhibit steady state conditions even for electron densities of 10^{21}cm^{-3} .

This intuitive analysis was subjected to a detailed test using a rate equation model of an argon plasma.⁽²⁷⁾ Argon was chosen because recent interest in using it as a diagnostic fill gas in laser fusion experiments using glass microballoons has produced improved cross section data for many of the ionization stages and their excited states. Conclusions drawn from this calculation should apply with little change to corresponding stages of elements nearby in the periodic table.

The ionization dynamics of a small volume of plasma were studied by numerically solving a set of coupled equations for the electron and ion thermal energy densities together with the population densities of the ground state of the relevant ionization stages.⁽²⁸⁾ Collisional excitation rates using state-of-the-art cross sections for the 3s, 3p, and 3d states of carbon-like ArXIII were calculated along with radiative decay and collisional mixing rates.⁽²⁹⁾ In addition, these excited states were collisionally and radiatively coupled to the ArXIV ground state. The initial plasma in the volume element is taken to be cold and to have an ion density of $5.5 \times 10^9\text{cm}^{-3}$.

Incident laser flux rapidly heats the electrons and the subsequent ionization is computed. A plasma volume element of 68μ radius was chosen with an initial, uniform electron temperature of approximately 1.0ev. An absorption of 0.55 joule of laser energy in this volume in 100 psec raises the electron temperature to approximately 600ev and, though ionization, produces an electron density of 10^{21}cm^{-3} . (A plasma scale length of $20\mu\text{m}$

in a line focus geometry $100\mu\text{m} \times 1000\mu\text{m}$ has approximately the same volume as the calculated case and is well matched to our experimental conditions.)

In Fig. 5 the densities of the important stages of ionization are shown as a function of time as computed with this model. Under the assumed conditions the density of carbon-like ions reaches a maximum near the peak of the incident laser heating pulse. This insures strong collisional excitation of carbon-like excited states.

A partial energy level diagram of the carbon-like stage is given in Fig. 6 showing the predicted laser transition and some of the coupling processes assumed in the calculation. The 3p state is pumped directly from the ground state and from collisional mixing with the 3d state. Collisional excitation of the 3s state from the ground state also occurs. However, a transient inversion is created between the 3p and 3s states due to the more rapid radiative decay of the 3s state, even when collisional mixing on the transition is accounted for.

In Fig. 7 the calculated gain coefficient is plotted as a function of time along with the computed time history of the electron temperature. A gain coefficient in excess of 100cm^{-1} is predicted for a time of approximately 100 psec. A useful active length of 3cm of plasma filament could thus be excited without the need for a travelling wave pump. Of course, the calculation does not include any provision for gain saturation which might occur due to strong amplification of spontaneous emission along the axis of such a plasma.

The conditions modelled here translate into readily realizable conditions in the laboratory. The plasma volume element studied closely approximates reasonable line focus dimensions and measured plasma scale lengths. The absorbed energy is consistent with the delivery of approxi-

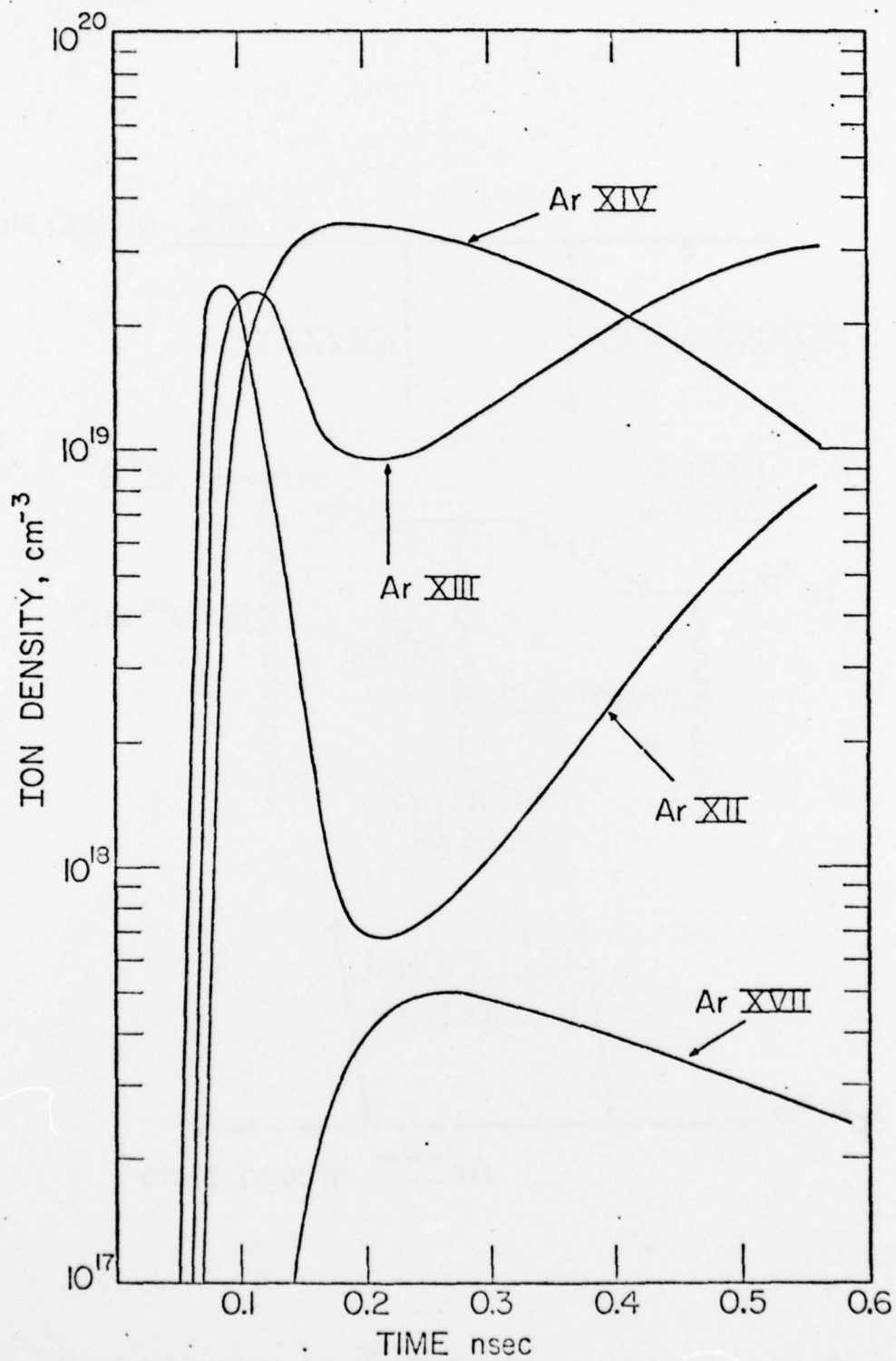


Fig. 5 Computed time history of ion densities of a plasma formed by absorption of 0.55 joule of 1.06 μ m laser energy in 10⁻¹⁰ sec by a solid argon target. A plasma volume element 68 μ m in radius is assumed.

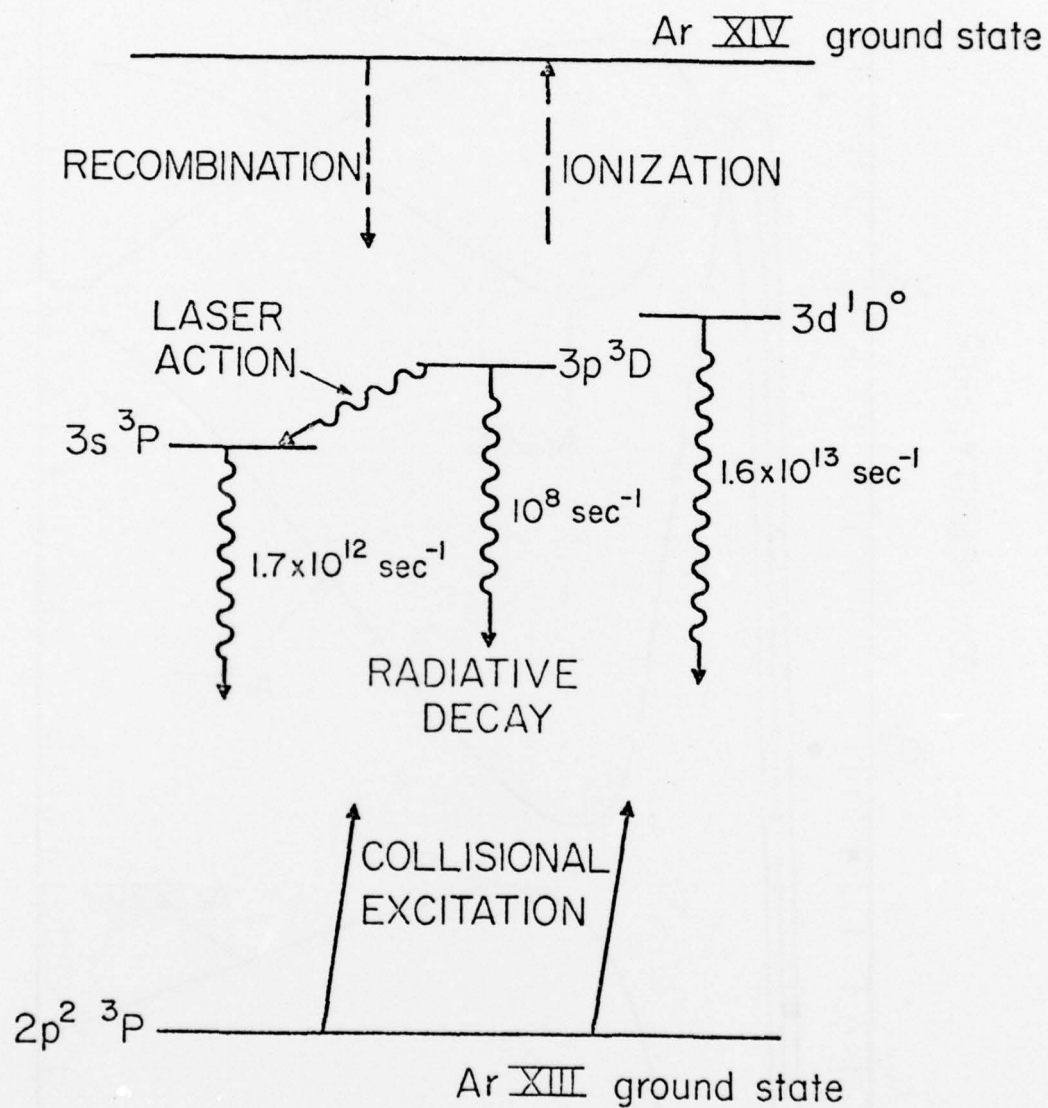


Fig. 6 Partial energy level diagram of carbon-like argon.

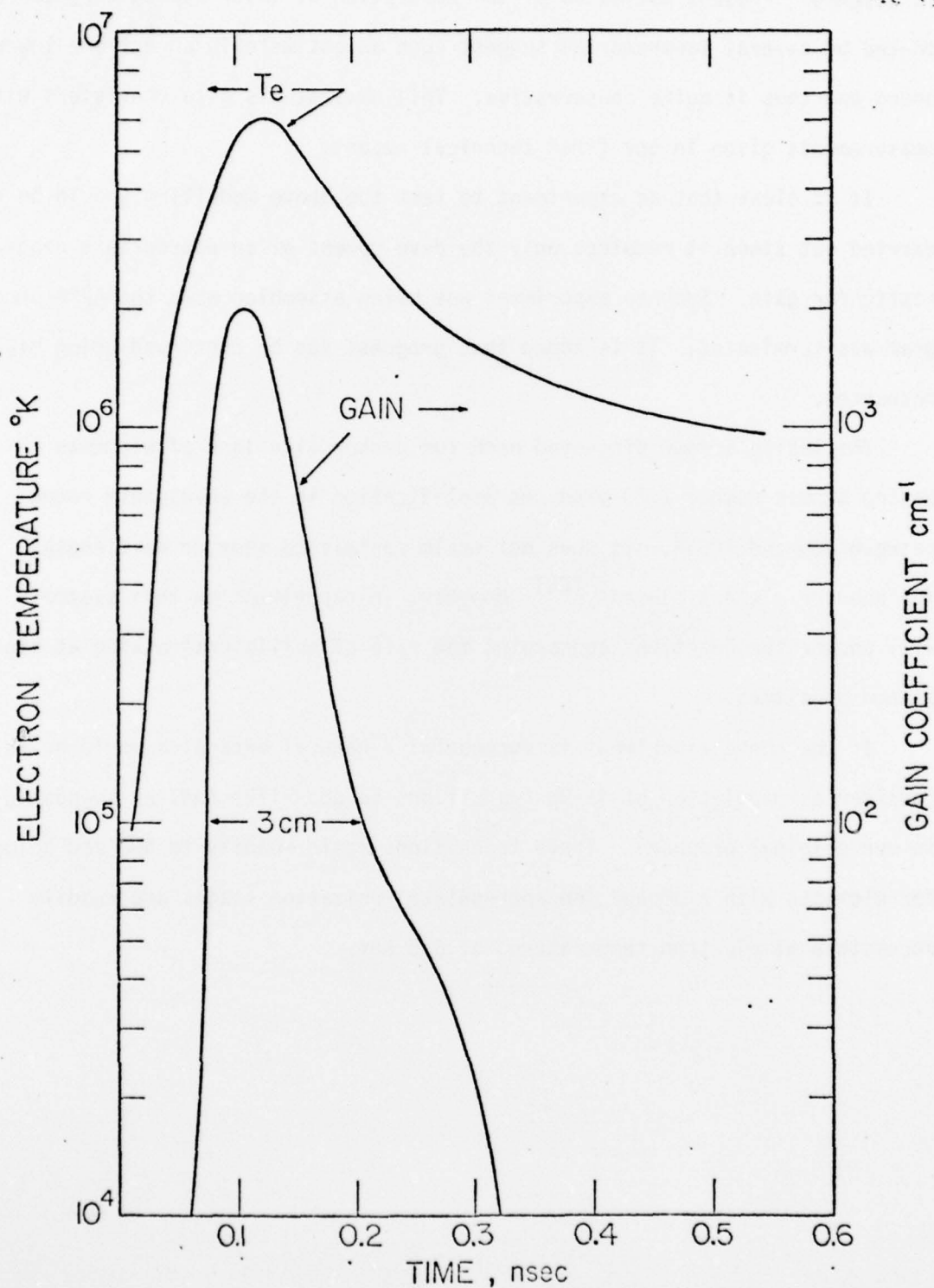


Fig. 7 Time history of the electron temperature and the gain coefficient for carbon-like ions of argon produced in the vicinity of critical electron density of a 1.06μ laser plasma.

mately 10 joules of laser energy in 10^{-10} sec, if a 5% absorption is assumed. Present estimates of the absorption of laser heated targets reported by several laboratories suggest such an estimate is an extreme lower bound and thus is quite conservative. This estimate is also consistent with measurements given in our first technical report.

It is clear that an experiment to test the above modelling should be carried out since it requires only the development of an appropriate diagnostic for gain. Such an experiment was being assembled when the ARPA program was terminated. It is hoped that progress can be continued using other resources.

The lasing scheme discussed here for carbon-like ions of elements having atomic number $Z \geq 20$ produces amplification in the wavelength range between 500 and 1000\AA . It does not scale rapidly to shorter wavelengths for heavier element plasmas.⁽²⁵⁾ However, an experiment on this system will answer the questions concerning the rate of collisional mixing at high plasma densities.

If the above experiment is successful a natural extension would be to consider extrapolation of $4s \rightarrow 3p$ transitions in neon-like ions as suggested in our original proposal. These transitions scale readily to 50\AA and below for elements with $Z \leq 30$ and the appropriate ionization stages are readily accessible at electron temperatures of 0.5 Kev.

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